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(71) Applicant: **Lg Chem, Ltd.**
Seoul 07336 (KR)

(72) Inventors:
- **LEE, Jeongbin**
  Daejeon 34122 (KR)
- **PARK, Eunju**
  Daejeon 34122 (KR)
- **LEE, Ki Jae**
  Daejeon 34122 (KR)
- **BAE, Jungmoon**
  Daejeon 34122 (KR)
- **HAN, Joong Jin**
  Daejeon 34122 (KR)
- **HONG, Mooho**
  Daejeon 34122 (KR)

(74) Representative: **Goddar, Heinz J.**
Boehmert & Boehmert
Anwaltspartnerschaft mbB
Pettenkoferstrasse 22
80336 München (DE)

(54) **MONOMER COMPOSITION FOR SYNTHESIS OF RECYCLED PLASTIC, METHOD FOR PREPARING SAME, AND RECYCLED PLASTIC AND MOLDED PRODUCT USING SAME**

(57) The present invention relates to a monomer composition for synthesizing recycled plastic which contains a high-purity aromatic diol compound recovered through recycling by chemical decomposition of a polycarbonate-based resin, a method for preparing the same, and a recycled plastic, and molded product using the same.

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Description

[TECHNICAL FIELD]

CROSS-REFERENCE TO RELATED APPLICATION(S)

[0001] This application claims the benefit of Korean Patent Application No. 10-2021-0122001 filed on September 13, 2021, Korean Patent Application No. 10-2021-0122002 filed on September 13, 2021, Korean Patent Application No. 10-2021-0122003 filed on September 13, 2021, Korean Patent Application No. 10-2021-0122004 filed on September 13, 2021, Korean Patent Application No. 10-2021-0128892 filed on September 29, 2021, and Korean Patent Application No. 10-2021-0136153 filed on October 13, 2021 in the Korean Intellectual Property Office, the entire contents of which are incorporated herein by reference.

[0002] The present invention relates to a monomer composition for synthesizing recycled plastic that contains a high-purity aromatic diol compound recovered through recycling by chemical decomposition of a polycarbonate-based resin, a method for preparing the same, and a recycled plastic and molded product using the same.

[BACKGROUND ART]

[0003] Polycarbonate is a thermoplastic polymer and is a plastic having excellent characteristics such as excellent transparency, ductility, and relatively low manufacturing cost.

[0004] Although polycarbonate is widely used for various purposes, environmental and health concerns during waste treatment have been continuously raised.

[0005] Currently, a physical recycling method is carried out, but in this case, a problem accompanying the deterioration of the quality occurs, and thus, research on the chemical recycling of polycarbonate is underway.

[0006] Chemical decomposition of polycarbonate refers to obtaining an aromatic diol compound as a monomer (e.g., bisphenol A (BPA)) through decomposition of polycarbonate, and then utilizing it again in polymerization to obtain a high-purity polycarbonate.

[0007] For such a chemical decomposition, thermal decomposition, hydrolysis, and alcohol decomposition are typically known. Among these, the most common method is alcohol decomposition using a base catalyst, but in the case of methanol decomposition, there is a problem that methanol is used which is harmful to the human body, and in the case of ethanol, there is a problem that high temperature and high pressure conditions are required and the yield is not high.

[0008] In addition, although an alcohol decomposition method using an organic catalyst is known, it is disadvantageous in terms of economics.

[DETAILED DESCRIPTION OF THE INVENTION]

[Technical Problem]

[0009] It is an object of the present invention to provide a monomer composition for synthesizing recycled plastic that can secure a high-purity aromatic diol compound recovered through recycling by chemical decomposition of a polycarbonate-based resin.

[0010] It is another object of the present invention to provide a method for preparing the monomer composition for synthesizing recycled plastic, and a recycled plastic, and molded product using the monomer composition for synthesizing recycled plastic.

[Technical Solution]

[0011] In order to achieve the above object, provided herein is a monomer composition for synthesizing recycled plastic, which comprises an aromatic diol compound, wherein diethyl carbonate is obtained as a by-product, wherein the recovery rate of diethyl carbonate, which is a by-product, according to the following mathematical formula is 82% or more, and wherein the monomer composition for synthesizing recycled plastic is recovered from a polycarbonate-based resin.

[0012] Also provided herein is a method for preparing a monomer composition for synthesizing recycled plastic, the method comprising the steps of: depolymerizing a polycarbonate-based resin; adding a first adsorbent to the depolymerization reaction product to perform an adsorption purification and then removing the first adsorbent; separating a carbonate precursor from the depolymerization reaction product; and purifying the depolymerization reaction product from which the carbonate precursor has been separated, wherein the step of purifying the depolymerization reaction product from which the carbonate precursor has been separated comprises: a step of washing the depolymerization

reaction product from which the carbonate precursor has been separated; and a step of adding a second adsorbent to the depolymerization reaction product from which the carbonate precursor has been separated to perform an adsorption purification and then removing the second adsorbent.

[0013] Further provided herein is a recycled plastic comprising a reaction product of the monomer composition for synthesizing recycled plastic and a comonomer.

[0014] Further provided herein is a molded product comprising the recycled plastic.

[0015] Below, a monomer composition for synthesizing recycled plastic, a method for preparing the same, and a recycled plastic, and molded product using the same according to specific embodiments of the present invention will be described in more detail.

[0016] Unless explicitly stated herein, the technical terms used herein are for the purpose of describing specific embodiments only and is not intended to limit the scope of the invention.

[0017] The singular forms "a," "an" and "the" used herein are intended to include plural forms, unless the context clearly indicates otherwise.

[0018] It should be understood that the terms "comprise," "include", "have", etc. are used herein to specify the presence of stated feature, region, integer, step, action, element and/or component, but do not preclude the presence or addition of one or more other feature, region, integer, step, action, element, component and/or group.

[0019] Further, the terms including ordinal numbers such as "a first", "a second", etc. are used only for the purpose of distinguishing one component from another component, and are not limited by the ordinal numbers. For instance, a first component may be referred to as a second component, or similarly, the second component may be referred to as the first component, without departing from the scope of the present invention.

1. Monomer Composition for Synthesizing Recycled Plastic

[0020] According to one embodiment of the present invention, there can be provided a monomer composition for synthesizing recycled plastic, which comprises an aromatic diol compound, wherein diethyl carbonate is obtained as a by-product, wherein the recovery rate of diethyl carbonate, which is a by-product, according to the following mathematical formula is 82% or more, and wherein the monomer composition for synthesizing recycled plastic is recovered from a polycarbonate-based resin.

[Mathematical Formula]

$$\text{Recovery rate of diethyl carbonate} = \{(\text{Number of moles of diethyl carbonate recovered}) / (\text{Number of moles of diethyl carbonate in entire by-product mixture})\} \times 100.$$

[0021] The present inventors have found through experiments that although the monomer composition for synthesizing recycled plastics of the one embodiment was recovered through recycling by chemical decomposition of the polycarbonate-based resin, the content of impurities other than an aromatic diol compound, which is the main recovery target, is remarkably reduced, thereby capable of realizing excellent physical properties in the synthesis of polycarbonate-based resins using the same, and completed the present invention.

[0022] In particular, along with the monomer composition (first composition) for synthesizing recycled plastics of the one embodiment, the monomer composition (second composition) for synthesizing recycled plastic which comprises diethyl carbonate wherein the diethyl carbonate is recovered from a polycarbonate-based resin, can be respectively simultaneously obtained in a method for preparing a monomer composition for synthesizing recycled plastics, which will be described later.

[0023] That is, the present invention can have technical features that a first composition comprising an aromatic diol compound is obtained with high purity through recycling by chemical decomposition of polycarbonate-based resin, and at the same time, a second composition comprising diethyl carbonate as a by-product with a high added value can also be obtained.

[0024] Specifically, the monomer composition for synthesizing recycled plastics of the one embodiment is characterized by being recovered from a polycarbonate-based resin. That is, this means that recovery is performed from the polycarbonate-based resin in order to obtain the monomer composition for synthesizing recycled plastics of the one embodiment, and as a result, the monomer composition for synthesizing recycled plastics containing the aromatic diol compound is obtained together.

[0025] The polycarbonate-based resin is meant to include both a homopolymer and a copolymer containing a polycarbonate repeating unit, and collectively refers to a reaction product obtained through a polymerization reaction or a

copolymerization reaction of a monomer containing an aromatic diol compound and a carbonate precursor. When it contains one carbonate repeating unit obtained by using only one aromatic diol compound and one carbonate precursor, a homopolymer can be synthesized. In addition, when one aromatic diol compound and two or more carbonate precursors are used as the monomer, or two or more aromatic diol compounds and one carbonate precursor are used, or one or more other diols is used in addition to the one aromatic diol compound and the one carbonate precursor to contain two or more carbonates, a copolymer can be synthesized. The homopolymer or copolymer can include all of low-molecular compounds, oligomers, and polymers depending on the molecular weight range.

[0026] Further, the monomer composition for synthesizing recycled plastics of the one embodiment may include an aromatic diol compound. Specific examples of the aromatic diol compound include bis(4-hydroxyphenyl)methane, bis(4-hydroxyphenyl)ether, bis(4-hydroxyphenyl)sulfone, bis(4-hydroxyphenyl)sulfoxide, bis(4-hydroxyphenyl)sulfide, bis(4-hydroxyphenyl)ketone, 1,1-bis(4-hydroxyphenyl)ethane, 2,2-bis(4-hydroxyphenyl)propane (bisphenol A), 2,2-bis(4-hydroxyphenyl)butane, 1,1-bis(4-hydroxyphenyl)cyclohexane (bisphenol Z), 2,2-bis(4-hydroxy-3,5-dibromophenyl)propane, 2,2-bis(4-hydroxy-3,5-dichlorophenyl)propane, 2,2-bis(4-hydroxy-3-bromophenyl)propane, 2,2-bis(4-hydroxy-3-chlorophenyl)propane, 2,2-bis(4-hydroxy-3 -methylphenyl)propane, 2,2-bis(4-hydroxy-3,5-dimethylphenyl)propane, 1,1-bis(4-hydroxyphenyl)-1-phenylethane, or a mixture of two or more thereof, and the like. Preferably, the aromatic diol compound of the monomer composition for synthesizing recycled plastics of the one embodiment may be 2,2-bis(4-hydroxyphenyl)propane (bisphenol A).

[0027] The aromatic diol compound is characterized by being recovered from the polycarbonate-based resin used for recovering the monomer composition for synthesizing the recycled plastic. That is, this means that recovery is performed from the polycarbonate-based resin in order to obtain the monomer composition for synthesizing recycled plastics of the one embodiment, and as a result, an aromatic diol compound is also obtained together. Therefore, apart from the recovery from the polycarbonate-based resin in order to prepare the monomer composition for synthesizing recycled plastics of the one embodiment, the case where a novel aromatic diol compound is added from the outside is not included in the category of aromatic diol compound of the present invention.

[0028] Specifically, "recovered from the polycarbonate-based resin" means being obtained through a depolymerization reaction of the polycarbonate-based resin. The depolymerization reaction can be carried out under acidic, neutral or basic conditions, and particularly, the depolymerization reaction can proceed under basic (alkaline) conditions. Particularly, the depolymerization reaction can be preferably carried out in the presence of an ethanol solvent, as will be described later.

[0029] Meanwhile, the monomer composition for synthesizing recycled plastics of the one embodiment may have a color coordinate b* value of 2.5 or less, or 0.1 or more, or 0.1 to 2.5, or 1.0 to 2.5. Also, the monomer composition for synthesizing recycled plastics of the one embodiment may have a color coordinate L* of 96.1 or more, or 100 or less, or 96.1 to 100, or 96.1 to 99. Further, the monomer composition for synthesizing recycled plastics of the one embodiment may have a color coordinate a* of 0.4 or less, or -0.1 or more, or -0.1 to 0.4, or -0.07 to 0.4.

[0030] As used herein, the "color coordinates" means coordinates in the CIE Lab color space, which are color values defined by CIE (Commossion International de l'Eclairage), and an arbitrary position in the CIE color space may be represented by three coordinate values, i.e., L*, a*, and b*.

[0031] Here, the L* value represents brightness, when L*=0, it represents black, and when L*=100, it represents white. In addition, the a* value represents a color having a corresponding color coordinate that leans toward one of pure red and pure green, and the b* value represents a color having a corresponding color coordinate that leans toward one of pure yellow and pure blue.

[0032] Specifically, the a* value is in the range of -a to +a. A maximum value of a* (a* max) represents pure red, and a minimum value of a* (a* min) represents pure green. Further, the b* value is in the range of -b to +b. A maximum value of b* (b* max) represents pure yellow, and a minimum value of b* (b* min) represents pure blue. For example, a negative b* value represents a color leaning toward pure blue, and a positive b* value represents color leaning toward pure yellow. When comparing b*=50 with b*=80, b*=80 is closer to pure yellow than b*=50.

[0033] When the color coordinate b* value of the monomer composition for synthesizing recycled plastics of the one embodiment excessively increases to more than 2.5, or the color coordinate L* value excessively decreases to less than 96.1, or the color coordinate a* value excessively increases to more than 0.4, the monomer composition for synthesizing recycled plastics of the one embodiment deteriorates in the color characteristics. Examples of the method for measuring the color coordinates L*, a*, b* values of the monomer composition for synthesizing recycled plastics of the one embodiment are not particularly limited, and various color characteristic measurement methods in the field of plastics can be applied without limitation.

[0034] However, the color coordinates L*, a*, and b* values of the monomer composition for synthesizing recycled plastics of the one embodiment can be measured in reflection mode using HunterLab UltraScan PRO Spectrophotometer as an example.

[0035] Meanwhile, the monomer composition for synthesizing recycled plastics of the one embodiment may have an aromatic diol compound purity of 96% or more, or 100% or less, or 96% to 100%.

[0036] Examples of the method for measuring the purity of the aromatic diol compound of the monomer composition for synthesizing recycled plastics of the one embodiment are not particularly limited, and for example, ^{1}H NMR, ICP-MS analysis, HPLC analysis, UPLC analysis, etc. can be used without limitation. As for the specific methods, conditions, equipment, etc. of the NMR, ICP-MS, HPLC, and UPLC, various well-known contents can be applied without limitation.

[0037] An example of a method for measuring the purity of the aromatic diol compound of the monomer composition for synthesizing recycled plastics of the one embodiment is follows. 1 wt% of the monomer composition for synthesizing recycled plastics of the one embodiment was dissolved in acetonitrile (ACN) solvent under normal pressure and 20 to 30°C conditions, and then the purity of bisphenol A (BPA) was analyzed by ultraperformance liquid chromatography (UPLC) on a Waters HPLC system using ACQUITY UPLC®BEH C18 1.7 μm (2.1*50mm column).

[0038] As described above, in the monomer composition for synthesizing recycled plastics of the one embodiment, the purity of the aromatic diol compound, which is the main recovery target material, is greatly increased to 96% or more, and other impurities are minimized, thereby capable of achieving excellent physical properties when synthesizing a polycarbonate-based resin using the same.

[0039] Meanwhile, in the monomer composition for synthesizing recycled plastics of the one embodiment, diethyl carbonate may be obtained as a by-product. The diethyl carbonate is characterized by being recovered from the polycarbonate-based resin used for recovering the monomer composition for recycling plastic synthesis of the one embodiment.

[0040] That is, this means that recovery is performed from the polycarbonate-based resin in order to obtain the monomer composition for synthesizing recycled plastics of the one embodiment, and as a result, diethyl carbonate is also obtained together. Therefore, apart from the recovery from the polycarbonate-based resin in order to prepare the monomer composition for synthesizing recycled plastics of the one embodiment, the case where a novel diethyl carbonate is added from the outside is not included in the category of diethyl carbonate of the present invention.

[0041] Specifically, "recovered from the polycarbonate-based resin" means being obtained through a depolymerization reaction of the polycarbonate-based resin. The depolymerization reaction can be carried out under acidic, neutral or basic conditions, and particularly, the depolymerization reaction can proceed under basic (alkaline) conditions. Particularly, the depolymerization reaction can be preferably carried out in the presence of an ethanol solvent, as will be described later.

[0042] Since the main recovery target material in the monomer composition for synthesizing recycled plastics of the one embodiment is an aromatic diol compound, the diethyl carbonate can be separately separated and recovered as the by-product from the monomer composition for synthesizing recycled plastics of the one embodiment.

[0043] Meanwhile, in the monomer composition for synthesizing recycled plastics of the one embodiment, the recovery rate of diethyl carbonate, which is a by-product, according to the mathematical formula may be 82% or more, or 83% or more, or 84% or more, or 85% or more, or 100% or less, or 82% to 100%, or 83% to 100%, or 84% to 100%, or 85% to 100%.

[0044] The example of a method for measuring the recovery rate of diethyl carbonate, which is a by-product of the monomer composition for synthesizing recycled plastics of the one embodiment, is not particularly limited, and for example, ^{1}H NMR, ICP-MS analysis, HPLC analysis, UPLC analysis, etc. can be used without limitation. As for the specific methods, conditions, equipment, etc. of the NMR, ICP-MS, HPLC, and UPLC, various well-known contents can be applied without limitation.

[0045] As described above, in the monomer composition for synthesizing recycled plastics of the one embodiment, the recovery rate of diethyl carbonate, which is a by-product, according to the mathematical formula is extremely increased to 82% or more, so that diethyl carbonate with high added value can be recovered with high efficiency to increase economic efficiency, and the efficiency can be increased in the synthesis of various plastics (polycarbonate-based resin, polyurethane resin, epoxy resin, etc.) using the same.

[0046] The monomer composition for synthesizing recycled plastics of the one embodiment can be used as a raw material for preparing various recycled plastics (e.g., polycarbonate (PC)) which will be described later.

[0047] The monomer composition for synthesizing recycled plastics of the one embodiment may further include small amounts of other additives and solvents. Specific types of the additives or solvents are not particularly limited, and various materials widely used in the process of recovering the aromatic diol compound by a depolymerization of the polycarbonate-based resin can be applied without limitation.

[0048] The monomer composition for synthesizing recycled plastics of the one embodiment can be obtained by a method for preparing a monomer composition for synthesizing recycled plastics, which will be described later. That is, the monomer composition for synthesizing recycled plastics of one embodiment corresponds to the result obtained through various processes of filtration, purification, washing, and drying in order to secure only the aromatic diol compound, which is the main recovery target material, with high purity after the depolymerization reaction of the polycarbonate-based resin.

2. Method for preparing a monomer composition for synthesizing recycled plastic

[0049] According to another embodiment of the invention, there can be provided a method for preparing a monomer composition for synthesizing recycled plastic, the method comprising the steps of: depolymerizing a polycarbonate-based resin; adding a first adsorbent to the depolymerization reaction product to perform an adsorption purification and then removing the first adsorbent; separating a carbonate precursor from the depolymerization reaction product; and purifying the depolymerization reaction product from which the carbonate precursor has been separated, wherein the step of purifying the depolymerization reaction product from which the carbonate precursor has been separated comprises: a step of washing the depolymerization reaction product from which the carbonate precursor has been separated; and a step of adding a second adsorbent to the depolymerization reaction product from which the carbonate precursor has been separated to perform an adsorption purification and then removing the second adsorbent.

[0050] The present inventors have confirmed through experiments that similarly to the method for preparing the monomer composition for synthesizing recycled plastic of the other embodiments, in the process of recycling polycarbonate-based resin by chemical decomposition, a first adsorption purification step using a first adsorbent and a second adsorption purification process using a second adsorbent are applied together and at the same time, the washing step is included in the purification process, whereby in the present invention, the aromatic diol compound, which is the main synthesis target material, can be secured with high purity, and the content of impurities other than the aromatic diol compound can be significantly reduced, and completed the present invention.

[0051] Specifically, the method for preparing the monomer composition for synthesizing recycled plastics of the other embodiment may include a pretreatment step of the polycarbonate-based resin, before the step of depolymerizing the polycarbonate-based resin.

[0052] The polycarbonate-based resin is meant to include both a homopolymer and a copolymer containing a polycarbonate repeating unit, and collectively refers to a reaction product obtained through a polymerization reaction or a copolymerization reaction of a monomer containing an aromatic diol compound and a carbonate precursor. When it contains one carbonate repeating unit obtained by using only one aromatic diol compound and one carbonate precursor, a homopolymer can be synthesized. In addition, when one aromatic diol compound and two or more carbonate precursors are used as the monomer, or two or more aromatic diol compounds and one carbonate precursor are used, or one or more other diols is used in addition to the one aromatic diol compound and the one carbonate precursor to contain two or more carbonates, a copolymer can be synthesized. The homopolymer or copolymer can include all of low-molecular compounds, oligomers, and polymers depending on the molecular weight range.

[0053] The polycarbonate-based resin can be applied regardless of various forms and types, such as a novel polycarbonate-based resin produced through synthesis, a recycled polycarbonate-based resin produced through a regeneration process, or polycarbonate-based resin waste.

[0054] A pretreatment step of the polycarbonate-based resin is carried out, thereby capable of increasing the efficiency of the process of recovering the aromatic diol compound and the carbonate precursor from the polycarbonate-based resin. Examples of the pretreatment process may include washing, drying, grinding, glycol decomposition, and the like. The specific method of each pretreatment process is not limited, and various methods widely used in the process of recovering the aromatic diol compound and the carbonate precursor from the polycarbonate-based resin can be applied without limitation.

[0055] However, an example of the pretreatment process may include a method of dissolving the polycarbonate-based resin in an organic solvent and then filtering it. The organic solvent may include tetrahydrofuran, toluene, methylene chloride, chloroform, dimethyl carbonate, ethylmethyl carbonate, diethyl carbonate, dipropyl carbonate, or a mixture of two or more thereof.

[0056] Meanwhile, the method for preparing the monomer composition for synthesizing recycled plastics of the other embodiment may include a step of depolymerizing the polycarbonate-based resin.

[0057] During the depolymerization reaction of the polycarbonate-based resin, the depolymerization reaction may be carried out under acidic, neutral or basic conditions, and particularly, the depolymerization reaction may be carried out under basic (alkali) conditions. The type of the base is not particularly limited, and examples thereof include sodium hydroxide (NaOH) or potassium hydroxide (KOH). The base is a base catalyst acting as a catalyst, and has the economic advantages over organic catalysts, which are mainly used under mild conditions.

[0058] During the depolymerization reaction of the polycarbonate-based resin, the depolymerization reaction may be carried out by reacting a base in an amount of 0.5 mol or less, or 0.4 moles or less, or 0.3 moles or less, or 0.1 moles or more, or 0.2 mole or more, or 0.1 mole to 0.5 mole, or 0.1 mole to 0.4 mole, or 0.1 mole to 0.3 mole, or 0.2 mole to 0.5 mole, or 0.2 mole to 0.4 mole, or 0.2 mole to 0.3 mole relative to 1 mole of polycarbonate-based resin. When the polycarbonate-based resin is reacted with a base in an amount of more than 0.5 mol relative to 1 mole of the polycarbonate-based resin during depolymerization of the polycarbonate-based resin, it is limited that impurities increase due to the effect of increasing the amount of alkali salt generated, so the purity of the target recovery material is reduced, and the economic efficiency of the catalytic reaction is reduced.

[0059] Further, the depolymerization reaction of the polycarbonate-based resin can be carried out in the presence of a solvent including ethanol. The present invention can stably obtain bisphenol A, which is a high-purity monomer, by decomposing a polycarbonate-based resin with a solvent including ethanol, and has the advantage that diethyl carbonate having a high added value can be further obtained as a reaction by-product.

[0060] The content of the ethanol may be 5 to 15 moles, or 8 to 13 moles relative to 1 mole of the polycarbonate-based resin. Since the ethanol has good solubility in bisphenol A, ethanol within the above range should be essentially contained. When the content of the ethanol is excessively reduced to less than 5 moles relative to 1 mole of the polycarbonate-based resin, it is difficult to sufficiently progress the alcohol decomposition of polycarbonate-based resin. On the other hand, when the content of ethanol is excessively increased to more than 15 moles relative to 1 mole of the polycarbonate-based resin, the economics of the process can be reduced due to excessive use of alcohol.

[0061] The solvent in which the depolymerization reaction of the polycarbonate-based resin proceeds may further include, in addition to ethanol, at least one organic solvent selected from the group consisting of tetrahydrofuran, toluene, methylene chloride, chloroform, dimethyl carbonate, ethylmethyl carbonate, diethyl carbonate, and dipropyl carbonate.

[0062] The organic solvent may include tetrahydrofuran, toluene, methylene chloride, chloroform, dimethyl carbonate, ethylmethyl carbonate, diethyl carbonate, dipropyl carbonate, or a mixture of two or more thereof.

[0063] More preferably, methylene chloride can be used as the organic solvent. When methylene chloride is used as an organic solvent to be mixed with the ethanol, there is an advantage that the dissolution properties in polycarbonate can be improved and the reactivity can be enhance.

[0064] The content of the organic solvent may be 16 moles to 20 moles, or 16 moles to 18 moles relative to 1 mole of the polycarbonate-based resin. In addition, the content of the organic solvent may be 1.5 to 2 moles relative to 1 mole of ethanol. By mixing the polycarbonate-based resin, ethanol, and an organic solvent within the above range, there is an advantage that the depolymerization reaction of the polymer can proceed at a desired level.

[0065] Meanwhile, the temperature at which the depolymerization reaction of the polycarbonate-based resin proceeds is not particularly limited, but for example, the reaction may proceed at a temperature of 20°C to 100°C, or 50°C to 70°C. In addition, the depolymerization of the polycarbonate-based resin may proceed for 1 hour to 30 hours, or 4 hours to 6 hours.

[0066] Specifically, the conditions are mild process conditions relative to the conventional pressurizing/high temperature process, and by performing stirring under the above conditions, the process can be performed in a mild process as compared to the pressurizing/high temperature process. In particular, when stirring at 50°C to 70°C for 4 to 6 hours, there is an advantage of obtaining the most efficient results in terms of reproducibility and acceptability.

[0067] That is, according to the present invention, by adjusting the type and mixing amount of the mixed solvent and the type and content of the base catalyst without using an organic catalyst, there is the advantage that a high-purity aromatic diol compound (e.g., bisphenol A) can be obtained under mild conditions without using a pressure/high temperature process, and diethyl carbonate can be obtained as a by-product by using an ethanol solvent.

[0068] More specifically, the step of depolymerizing the polycarbonate-based resin may include a first step of dissolving the carbonate-based resin in an organic solvent; and a second step of adding and stirring a catalyst solution containing ethanol and a base. In the first and second steps, the contents of ethanol, organic solvent, base, and polycarbonate-based resin are the same as described above.

[0069] Meanwhile, the method for preparing the monomer composition for synthesizing recycled plastics of the other embodiment may further include a step of neutralizing the depolymerization reaction product with an acid, before the step of adding a first adsorbent to the depolymerization reaction product to perform an adsorption purification and then removing the first adsorbent.

[0070] For example, the alkali decomposition product of the polycarbonate-based resin includes an aromatic diol compound, or a salt thereof, but the main recovery target material of the present invention is an aromatic diol compound. Therefore, in the case of the salt of the aromatic diol compound obtained by the alkaline decomposition, it can be converted into an aromatic diol compound through an additional acid neutralization process. That is, when the depolymerization reaction of the polycarbonate-based resin is an alkaline decomposition, it can undergo a neutralization reaction step with an acid.

[0071] The acid used in the neutralization reaction may be a strong acid, for example, hydrochloric acid (HCl). Due to the neutralization reaction by the strong acid, the pH can satisfy 4 or less or less than 2 upon completion of the neutralization reaction. The temperature during the neutralization reaction can be adjusted to 25°C or more and 100°C or less.

Further, if necessary, after proceeding the neutralization reaction step of the depolymerization reaction product by an acid, a step of removing residual impurities through filtration or absorption can be further performed. Specifically, the aqueous layer and the organic layer can be separated, and the organic layer can be filtered through vacuum filtration to recover the liquid containing the aromatic diol compound.

[0072] Meanwhile, the method for preparing the monomer composition for synthesizing recycled plastics of the other embodiment may include a step of adding a first adsorbent to the depolymerization reaction product to perform an adsorption purification and then removing the first adsorbent. In the step of adding a first adsorbent to the depolymerization

reaction product to perform an adsorption purification and then removing the first adsorbent, An adsorbent can be brought into contact with the depolymerization reaction product.

[0073] Examples of the first adsorbent that can be used include activated carbon, charcoal, celite, or a mixture thereof. The activated carbon is a black carbon material having micropores produced by subjecting a raw material to a carbonization process at about 500°C and an activated carbon process at about 900°C, and examples thereof are not particularly limited, but for example, various activated carbons such as plant-based, coal-based, petroleum-based, waste-based activated carbons can be applied without limitation depending on the type of raw material.

[0074] In more specific examples, the plant-based activated carbon may include coconut activated carbon, wood activated carbon, and sawdust activated carbon. Further, the coal-based activated carbon may include lignite activated carbon, bituminous coal activated carbon, and anthracite activated carbon. Further, the petroleum-based activated carbon may include petroleum coke activated carbon and oil carbon activated carbon. Further, the waste-based activated carbon may include synthetic resin activated carbon and pulp activated carbon.

[0075] The first adsorbent may include at least one activated carbon selected from the group consisting of plant-based activated carbon, coal-based activated carbon, petroleum-based activated carbon, and waste-based activated carbon. That is, the first adsorbent may include plant-based activated carbon, coal-based activated carbon, petroleum-based activated carbon, waste-based activated carbon, or a mixture of two or more thereof.

[0076] More specifically, the first adsorbent may include at least one activated carbon selected from the group consisting of coconut activated carbon, lignite activated carbon, anthracite activated carbon, and bituminous coal activated carbon. That is, the first adsorbent may include coconut activated carbon, lignite activated carbon, anthracite activated carbon, bituminous coal activated carbon, or a mixture of two or more thereof.

[0077] Adsorption purification conditions by the first adsorbent are not particularly limited, and various well-known adsorption purification conditions can be used without limitation. However, for example, the addition amount of the adsorbent may be 40% to 60% by weight relative to the polycarbonate-based resin, and the adsorption time may be 0.1 hour to 5 hours, and the adsorption method may be a stirring adsorption or an adsorption tower for Lab.

[0078] Meanwhile, the method for preparing the monomer composition for synthesizing recycled plastics of the other embodiment may include a step of separating a carbonate precursor from the depolymerization reaction product. Thus, the separated carbonate precursor may include diethyl carbonate.

[0079] For example, the depolymerization reaction product of the polycarbonate-based resin includes an aromatic diol compound or a salt thereof and a carbonate precursor. The contents related to the aromatic diol compound and the carbonate precursor include all the contents described above in the one embodiment.

[0080] In the step of separating a carbonate precursor from the depolymerization reaction product, a reduced pressure distillation step of the depolymerization reaction product can be included. Examples of the reduced pressure distillation conditions are not particularly limited, but in a specific example, the depolymerization reaction product of the polycarbonate-based resin is pressurized under a pressure of 200 mbar to 300 mbar and a temperature of 20°C to 30°C, and depressurized under a pressure of 10 mbar to 50 mbar and a temperature of 20°C to 30°C and subjected to a low-temperature distillation.

[0081] The separated carbonate precursor can be recycled without a separate purification process, or can be recycled through separation and purification such as conventional extraction, adsorption, and drying, if necessary. Specific purification conditions are not particularly limited. As for the specific refining equipment and methods, various well-known purification techniques can be applied without limitation.

[0082] Meanwhile, the method for preparing the monomer composition for synthesizing recycled plastics of the other embodiment may include a step of purifying the depolymerization reaction product from which the carbonate precursor has been separated. Thereby, an aromatic diol compound, which is the main recovered material, is obtained, which corresponds to the monomer composition for synthesizing recycled plastic according to the one embodiment.

[0083] Specifically, the purification step of the depolymerization reaction product from which the carbonate precursor has been separated may include a step of washing the depolymerization reaction product from which the carbonate precursor has been separated; and a step of adding a second adsorbent to the depolymerization reaction product from which the carbonate precursor has been separated to perform an adsorption purification and then removing the second adsorbent.

[0084] In the purification step, the order of the washing step and the adsorption purification step is not particularly limited, and it is irrelevant to proceed in any order, but for example, the purification step may proceed in the order of the washing step; and the absorption purification step. The washing step; and the absorption purification step can proceed repeatedly at least once or more. As for the specific washing and adsorption equipment and methods, various well-known purification techniques can be applied without limitation.

[0085] Specifically, in the washing step of the depolymerization reaction product from which the carbonate precursor has been separated, the depolymerization reaction product from which the carbonate precursor has been separated may contain an aromatic diol compound. However, since various impurities remain during the recovery process of obtaining the aromatic diol compound, washing can proceed in order to sufficiently remove these impurities and secure

a high-purity aromatic diol compound.

[0086] Specifically, the washing step may include a step of washing with a solvent at a temperature of 10°C or more and 30°C or less, or 20°C or more and 30°C or less; and a step of washing with a solvent at a temperature of 40°C or more and 80°C or less, or 40°C or more and 60°C or less, or 45°C or more and 55°C or less. The temperature condition means the temperature inside the washing container at which washing with a solvent is performed. In order to maintain a high temperature deviating from a room temperature, various heating devices can be applied without limitation.

[0087] In the washing step, the step of washing with a solvent at a temperature of 10°C or more and 30°C or less may be performed first, and the step of washing with a solvent at a temperature of 40°C or more and 80°C or less may be performed later. Alternatively, the step of washing with a solvent at a temperature of 40°C or more and 80°C or less may be performed first, and the step of washing with a solvent at a temperature of 10°C or more and 30°C or less may be performed later.

[0088] More preferably, in the washing step, the step of washing with a solvent at a temperature of 10°C or more and 30°C or less may be performed first, and the step of washing with a solvent at a temperature of 40°C or more and 80°C or less may be performed later. Thereby, the corrosion of the reactor due to strong acid after the neutralization step can be minimized.

[0089] The step of washing with a solvent at a temperature of 10°C or more and 30°C or less; and the step of washing with a solvent at a temperature of 40°C or more and 80°C or less can be repeated at least once or more, respectively.

[0090] Further, if necessary, after proceeding the step of washing with a solvent at a temperature of 10°C or more and 30°C or less; and the step of washing with a solvent at a temperature of 40°C or more and 80°C or less, a step of removing the residual solvent through filtration may be further performed.

[0091] More specifically, the difference value between the temperature of the step of washing with a solvent at a temperature of 40°C or more and 80°C or less and the temperature of the step of washing with a solvent at a temperature of 10°C or more and 30°C or less may be 20°C or more and 50°C or less.

[0092] The difference value between the temperature of the step of washing with a solvent at a temperature of 40°C or more and 80°C or less and the temperature of the step of washing with a solvent at a temperature of 10°C or more and 30°C or less means a value obtained by subtracting the temperature of the step of washing with a solvent at a temperature of 10°C or more and 30°C or less from the temperature of the step of washing with a solvent at a temperature of 40°C or more and 80°C or less.

[0093] When the difference value between the temperature of the step of washing with a solvent at a temperature of 40°C or more and 80°C or less and the temperature of the step of washing with a solvent at a temperature of 10°C or more and 30°C or less decreases excessively to less than 20°C, it is difficult to sufficiently remove impurities.

[0094] When the difference value between the temperature of the step of washing with a solvent at a temperature of 40°C or more and 80°C or less and the temperature of the step of washing with a solvent at a temperature of 10°C or more and 30°C or less increases excessively to more than 50°C, severe conditions are formed to maintain extreme temperature conditions, which can reduce the efficiency of the process.

[0095] The solvent used in the washing step may include one of water, alcohol, and an organic solvent. As the organic solvent, tetrahydrofuran, toluene, methylene chloride, chloroform, dimethyl carbonate, ethylmethyl carbonate, diethyl carbonate, dipropyl carbonate, or a mixture of two or more thereof can be used.

[0096] The solvent used in the washing step can be used in a weight ratio of 1 part by weight or more and 30 parts by weight or less, or 1 part by weight or more and 10 parts by weight or less based on 1 part by weight of the polycarbonate-based resin used in the depolymerization reaction.

[0097] More specifically, the solvent in the step of washing with a solvent at a temperature of 10°C or more and 30°C or less may be an organic solvent. Preferably, methylene chloride can be used as the organic solvent. At this time, the organic solvent can be used in an amount of 1 part by weight or more and 10 parts by weight or less based on 1 part by weight of the polycarbonate-based resin.

[0098] Moreover, the solvent in the step of washing with a solvent at a temperature of 40°C or more and 80°C or less may be water. When water is used, impurities in the form of residual salts can be effectively removed. At this time, the solvent can be used in an amount of 1 part by weight or more and 10 parts by weight or less based on 1 part by weight of the polycarbonate-based resin.

[0099] Meanwhile, in the step of adding a second adsorbent to the depolymerization reaction product from which the carbonate precursor has been separated to perform an adsorption purification and then removing the second adsorbent, an adsorbent can be brought into contact with the depolymerization reaction product.

[0100] Examples of the second adsorbent that can be used include activated carbon, charcoal, celite, or a mixture thereof. The activated carbon is a black carbon material having micropores produced by subjecting a raw material to a carbonization process at about 500°C and an activated carbon process at about 900°C, and examples thereof are not particularly limited, but for example, various activated carbons such as plant-based, coal-based, petroleum-based, waste-based activated carbons can be applied without limitation depending on the type of raw material.

[0101] In more specific examples, the plant-based activated carbon may include coconut activated carbon, wood

activated carbon, and sawdust activated carbon. Also, the coal-based activated carbon may include lignite activated carbon, bituminous coal activated carbon, and anthracite activated carbon. Further, the petroleum-based activated carbon may include petroleum coke activated carbon and oil carbon activated carbon. Further, the waste-based activated carbon may include synthetic resin activated carbon and pulp activated carbon.

[0102] The second adsorbent may include at least one activated carbon selected from the group consisting of plant-based activated carbon, coal-based activated carbon, petroleum-based activated carbon, and waste-based activated carbon. That is, the second adsorbent may include plant-based activated carbon, coal-based activated carbon, petroleum-based activated carbon, waste-based activated carbon, or a mixture of two or more thereof.

[0103] More specifically, the second adsorbent may include at least one activated carbon selected from the group consisting of coconut activated carbon, lignite activated carbon, anthracite activated carbon, and bituminous coal activated carbon. That is, the second adsorbent may include coconut activated carbon, lignite activated carbon, anthracite activated carbon, bituminous coal activated carbon, or a mixture of two or more thereof.

[0104] As described above, in the method for preparing a monomer composition for synthesizing recycled plastics of the other embodiment, the first adsorption purification step with the first adsorbent and the second adsorption purification step with the second adsorbent are applied together during the depolymerization reaction in which the polycarbonate-based resin is recycled by chemical decomposition, whereby not only the aromatic diol compound, which is the main synthesis target material in the present invention, can be secured with high purity, but also the content of impurities other than the aromatic diol compound can be significantly reduced.

[0105] The first adsorbent and the second adsorbent are the same or different from each other, and each independently may include at least one activated carbon selected from the group consisting of plant-based activated carbon, coal-based activated carbon, petroleum-based activated carbon, and waste-based activated carbon. More specifically, the first adsorbent and the second adsorbent are the same or different from each other, and each independently may include at least one activated carbon selected from the group consisting of coconut activated carbon, lignite activated carbon, anthracite activated carbon, and bituminous coal activated carbon.

[0106] Examples of the weight ratio of adding the first adsorbent and the second adsorbent are not particularly limited, but as an example, the amount of the first adsorbent added may be 1 part by weight to 1000 parts by weight based on 100 parts by weight of the second adsorbent added.

[0107] The adsorption purification conditions by the second adsorbent are not particularly limited, and various well-known adsorption purification conditions can be used without limitation. However, for example, the addition amount of the adsorbent may be 40% by weight to 60% by weight relative to the polycarbonate-based resin, the adsorption time may be 0.1 hours to 5 hours, and the adsorption method may be a stirring adsorption or an adsorption tower for lab.

[0108] If necessary, the method may further include a step of adding a solvent to the depolymerization reaction product from which the carbonate precursor has been separated, before the step of adding a second adsorbent to the depolymerization reaction product from which the carbonate precursor has been separated to perform an adsorption purification and then removing the second adsorbent. Examples of the solvent include ethanol, and the ethanol may be added in a ratio of 1 mole to 20 moles, or 5 moles to 20 moles relative to 1 mole of the polycarbonate-based resin. Aromatic diol compound crystals included in the depolymerization reaction product from which the carbonate precursor has been separated can be redissolved in a solvent through the step of adding a solvent to the depolymerization reaction product in which the carbonate precursor has been separated.

[0109] Meanwhile, after the step of adding a second adsorbent to the depolymerization reaction product from which the carbonate precursor has been separated to perform an adsorption purification and then removing the second adsorbent, the purification step may further include a recrystallization step of the depolymerization reaction product from which the carbonate precursor has been separated.

[0110] In the recrystallization step of the depolymerization reaction product from which the carbonate precursor has been separated, a high-purity aromatic diol compound can be secured by sufficiently removing various impurities contained in the depolymerization product from which the carbonate precursor has been separated.

[0111] Specifically, the recrystallization step may include a step of adding water to the depolymerization reaction product from which the carbonate precursor has been separated to perform recrystallization. Through the step of adding water to the depolymerization reaction product from which the carbonate precursor has been separated to perform recrystallization, the solubility of the aromatic diol compound or its salt contained in the depolymerization reaction product is increased, and thus, crystals, or impurities interposed between crystals can be dissolved with a solvent to the maximum, and further, since the dissolved aromatic diol compound has poor solubility relative to impurities, it can be easily precipitated into aromatic diol compound crystals through the difference in solubility when the temperature is lowered subsequently.

[0112] More specifically, in the step of adding water to the depolymerization reaction product from which the carbonate precursor has been separated to perform recrystallization, 200 moles to 400 moles, or 250 moles to 350 moles of water can be used with respect to 1 mole of the polycarbonate-based resin. When the water is used in an excessively small amount, the temperature for dissolving the aromatic diol compound contained in the depolymerization reaction product

from which the carbonate precursor has been separated becomes too high, which thus deteriorates in the process efficiency, and it is difficult to remove impurities through recrystallization. On the other hand, when water is used in an excessive amount, the solubility of the aromatic diol compound contained in the depolymerization reaction product from which the carbonate precursor has been separated becomes too high, and thus, the yield of the aromatic diol compound recovered after recrystallization is reduced, and the process efficiency can be reduced due to the use of large amounts of solvent.

[0113] If necessary, after proceeding the recrystallization step of the depolymerization reaction product from which the carbonate precursor has been separated, a step of removing residual impurities through filtration or adsorption can be further performed.

[0114] In addition, if necessary, after the recrystallization step, the method may further include a drying step. The remaining solvent can be removed by the drying, and the specific drying conditions are not particularly limited, but for example, the drying can be performed at a temperature of 10°C to 100°C, or 10°C to 50°C. As for the specific drying equipment and method used in the drying, various well-known drying techniques can be applied without limitation.

3. Recycled Plastic

[0115] According to another embodiment of the invention, a recycled plastic comprising a reaction product of the monomer composition for synthesizing recycled plastic of the one embodiment and a comonomer can be provided.

[0116] The details of the monomer composition for synthesizing recycled plastic of the one embodiment include all the contents described above in the one embodiment and the other embodiment.

[0117] Examples corresponding to the recycled plastic are not particularly limited, and various plastics synthesized from aromatic diol compounds such as bisphenol A as a monomer can be applied without limitation, and a more specific example may be a polycarbonate-based resin.

[0118] The polycarbonate-based resin is meant to include both a homopolymer and a copolymer containing a polycarbonate repeating unit, and collectively refers to a reaction product obtained through a polymerization reaction or a copolymerization reaction of a monomer containing an aromatic diol compound and a carbonate precursor. When it contains one carbonate repeating unit obtained by using only one aromatic diol compound and one carbonate precursor, a homopolymer can be synthesized. In addition, when one aromatic diol compound and two or more carbonate precursors are used as the monomer, or two or more aromatic diol compounds and one carbonate precursor are used, or one or more other diols is used in addition to the one aromatic diol compound and the one carbonate precursor to contain two or more carbonates, a copolymer can be synthesized. The homopolymer or copolymer can include all of low-molecular compounds, oligomers, and polymers depending on the molecular weight range.

[0119] More specifically, in the recycled plastic containing the reaction product of the monomer composition for synthesizing the recycled plastic and the comonomer of the one embodiment, a carbonate precursor can be used as the comonomer. Specific examples of the carbonate precursor include phosgene, triphosgene, diphosgene, bromophosgene, dimethyl carbonate, diethyl carbonate, dibutyl carbonate, dicyclohexyl carbonate, diphenyl carbonate, ditolyl carbonate, bis(chlorophenyl)carbonate, m-cresyl carbonate, dinaphthyl carbonate, bis(diphenyl) carbonate or bishaloformate.

[0120] Examples of the reaction process of the monomer composition and the comonomer for synthesis are not particularly limited, and various well-known methods for preparing polycarbonate can be applied without limitation.

[0121] However, in one example of the polycarbonate preparation method, a polycarbonate preparation method including the step of polymerizing a composition containing a monomer composition for synthesizing recycled plastic and a comonomer can be used. At this time, the polymerization can be carried out by interfacial polymerization, and during interfacial polymerization, polymerization reaction is possible at normal pressure and low temperature, and the molecular weight is easy to control.

[0122] The polymerization temperature may be 0°C to 40°C, and the reaction time may be 10 minutes to 5 hours. In addition, the pH during the reaction may be maintained at 9 or more or 11 or more.

[0123] The solvent that can be used for the polymerization is not particularly limited as long as it is a solvent used for polymerization of polycarbonate in the art, and as an example, halogenated hydrocarbons such as methylene chloride and chlorobenzene can be used.

[0124] Moreover, the polymerization can be carried out in the presence of an acid binder. As the acid binder, an alkali metal hydroxide such as sodium hydroxide or potassium hydroxide, or an amine compound such as pyridine can be used.

[0125] Further, in order to adjust the molecular weight of the polycarbonate during the polymerization, polymerization can be performed in the presence of a molecular weight modifier. An alkylphenol having 1 to 20 carbon atoms may be used as the molecular weight modifier, and specific examples thereof include p-tert-butylphenol, p-cumylphenol, decylphenol, dodecylphenol, tetradecylphenol, hexadecylphenol, octadecylphenol, eicosylphenol, docosylphenol or triacontylphenol. The molecular weight modifier can be added before, during or after the initiation of polymerization, or after polymerization. The molecular weight modifier may be used in an amount of 0.01 to 10 parts by weight, or 0.1 to 6 parts

by weight, based on 100 parts by weight of the aromatic diol compound, and a desired molecular weight can be obtained within this range.

[0126] In addition, in order to promote the polymerization reaction, a reaction accelerator such as a tertiary amine compound, a quaternary ammonium compound, or a quaternary phosphonium compound, including triethylamine, tetra-n-butylammonium bromide, or tetra-n-butylphosphonium bromide can be further used.

4. Molded Product

[0127] According to another embodiment of the invention, a molded article comprising the recycled plastic of the other embodiment can be provided. The details of the recycled plastic includes all the contents described above in the other embodiments.

[0128] The molded article can be obtained by applying the recycled plastic to various known plastic molding methods without limitation. As an example of the molding method, injection molding, foam injection molding, blow molding, or extrusion molding may be mentioned.

[0129] Examples of the molded article are not particularly limited, and can be applied to various molded articles using plastic without limitation. Examples of the molded article include automobiles, electrical and electronic products, communication products, daily necessities, building materials, optical components, exterior materials, and the like.

[0130] The molded article may further include one or more additives selected from the group consisting of an antioxidant, a plasticizer, an antistatic agent, a nucleating agent, a flame retardant, a lubricant, an impact enhancer, an optical brightener, an ultraviolet absorber, a pigment and a dye, if necessary, in addition to the recycled plastic of the other embodiments,

[0131] An example of the manufacturing method of the molded article may include a step of mixing the recycled plastic of the other embodiment and an additive well using a mixer, extrusion-molding the mixture with an extruder to produce pellets, drying the pellets, and then injecting them with an injection molding machine.

[Advantageous Effects]

[0132] According to the present invention, a monomer composition for synthesizing recycled plastic that contains a high-purity aromatic diol compound recovered through recycling by chemical decomposition of a polycarbonate-based resin, a method for preparing the same, and a recycled plastic and molded product using the same can be provided.

[DETAILED DESCRIPTION OF THE EMBODIMENTS]

[0133] Hereinafter, the present invention will be explained in detail with reference to the following examples. However, these examples are for illustrative purposes only, and the scope of the present invention is not limited thereto.

<EXAMPLE>

Example 1

[0134]

(1. Decomposition step) 1 mol of Pretreated waste polycarbonate (PC) was dissolved in 17 mol of methylene chloride(MC), and then added together with 11 mol of ethanol (EtOH) and 0.25 mol of sodium hydroxide (NaOH) to a 3L high-pressure reactor, and the mixture was stirred at 60°C for 6 hours to proceed a PC depolymerization reaction.
(2. Neutralization stage) The depolymerization reaction product was cooled to 30°C or less, and then the product containing bisphenol A was neutralized using 0.25 mol of 10% hydrochloric acid(HCl) at 20~30°C.
(3-1. Purification-Adsorption step) After that, the product whose pH has been lowered to less than 2 was passed through an adsorption tower in which lignite activated carbon was added as a first adsorbent in a ratio of 50 wt.% relative to waste polycarbonate, and purified through adsorption.
(3-2. Purification-Distillation step) After that, the by-product diethyl carbonate(DEC) was separated and recovered through a low-temperature distillation reducing pressure from 250 mbar and 20~30°C to 30 mbar and 30°C.
(3-3. Purification-Washing step) After that, the residue from which diethyl carbonate (DEC) was removed was primarily washed using methylene chloride (MC) (1 time the mass of PC used) at 20 to 30°C, and vacuum filtered. The filtrate was secondarily washed using water (three times the mass of PC used) at a temperature of 50°C.
(4-1. Additional purification step - Redissolution step) 16.6 mol of Ethanol was added to the washed material and redissolved.
(4-2. Additional purification step - Adsorption step) After that, lignite activated carbon as the second adsorbent was

put into an adsorption tower at a ratio of 50 wt.% relative to waste polycarbonate, passed through it, and purified through adsorption.
(4-3. Additional purification step - Recrystallization step) After that, 300 mol of water was slowly added while stirring to recrystallize bisphenol A, and then filtered to obtain a solid.
(5. Drying step) After that, it was dried in a vacuum oven at 30~50°C to prepare a recycled bisphenol A monomer composition.

Examples 2 to 10

[0135] A recycled bisphenol A monomer composition and a recycled plastic were prepared in the same manner as in Example 1, except that in Example 1, the conditions of each step were changed as shown in Table 1 below.

[Table 1]

Category	(1.Decompositon step)				First adsorbent	Second adsorbent	(3-3. Purification-Washing step)		(4-1.Additional purification step-Redissolution step)	(4-3. Additional purification step-Recrystallization step)
	PC (eq)	MC (eq)	NaOH (eq)	EtOH (eq)			First washing	Second washing		
Example 1	1	17	0.25	11	Lignite activated carbon	Lignite activated carbon	MC / 20 ~ 30 °C	Water ~ r / 50 °C	EtOH / 16.6 mol eq	Water / 300 mol eq
Example 2	1	17	0.25	11	Lignite activated carbon	Lignite activated carbon	MC / 20 ~ 30 °C	Water / 20 ~ 30 °C	EtOH / 16.6 mol eq	Water / 300 mol eq
Example 3	1	17	0.25	11	Lignite activated carbon	Lignite activated carbon	MC / 20 ~ 30 °C	Water / 70 °C	EtOH / 16.6 mol eq	Water / 300 mol eq
Example 4	1	17	0.25	11	Lignite activated carbon	Lignite activated carbon	Water / 50 °C	MC / 20 ~ 30 °C	EtOH / 16.6 mol eq	Water / 300 mol eq
Example 5	1	20	0.25	11	Lignite activated carbon	Lignite activated carbon	MC / 20 ~ 30 °C	Water / 50 °C	EtOH / 16.6 mol eq	Water / 300 mol eq
Example 6	1	17	0.25	12.5	Lignite activated carbon	Lignite activated carbon	MC / 20 ~ 30 °C	Water / 50 °C	EtOH / 16.6 mol eq	Water / 300 mol eq
Example 7	1	17	0.25	8.5	Lignite activated carbon	Lignite activated carbon	MC / 20 ~ 30 °C	Water / 50 °C	EtOH / 16.6 mol eq	Water / 300 mol eq
Example 8	1	17	0.25	11	Lignite activated carbon	Lignite activated carbon	MC / 20 ~ 30 °C	Water / 50 °C	EtOH / 5.5 mol eq	Water / 300 mol eq
Example 9	1	17	0.25	11	Lignite activated carbon	Lignite activated carbon	MC / 20 ~ 30 °C	Water / 50 °C	EtOH / 11.1 mol eq	Water / 300 mol eq

(continued)

Category	(1.Decompositon step)				First adsorbent	Second adsorbent	(3-3. Purification-Washing step)		(4-1.Additional purification step-Redissolution step)	(4-3. Additional purification step-Recrystallization step)
	PC (eq)	MC (eq)	NaOH (eq)	EtOH (eq)			First washing	Second washing		
Example 10	1	17	0.25	11	Lignite activated carbon	Lignite activated carbon	MC / 20 ~ 30 °C	Water / 50 °C	EtOH / 16.6 mol eq	Water / 325 mol eq

<COMPARATIVE EXAMPLE>

Comparative Example 1

[0136] A recycled bisphenol A monomer composition was prepared in the same manner as in Example 1, except that (4-2. Additional purification step - Adsorption step) was not performed in Example 1.

Comparative Example 2

[0137] A recycled bisphenol A monomer composition was prepared in the same manner as in Example 1, except that (3-1. Purification-Adsorption step) was not performed in Example 1.

Comparative Example 3

[0138] A recycled bisphenol A monomer composition was prepared in the same manner as in Example 1, except that in the (3-3. Purification-Washing step) of Example 1, the residue from which diethyl carbonate (DEC) has been removed was subjected to a primary washing using methylene chloride (MC) (20 times the mass of PC used) at 20~30°C, and the secondary washing was not performed.

Comparative Example 4

[0139] A recycled bisphenol A monomer composition was prepared in the same manner as in Example 1, except that (3-3. Purification-Washing step) was not performed in Example 1.

<Experimental Example>

[0140] The physical properties of the recycled bisphenol A monomer composition or by-products obtained in Examples and Comparative Examples were measured by the following methods, and the results are shown in Table 2 below.

1. Purity

[0141] 1 wt% of Recycled bisphenol A monomer composition was dissolved in acetonitrile (ACN) solvent under normal pressure and 20 to 30°C conditions, and then the purity of bisphenol A (BPA) was analyzed by ultraperformance liquid chromatography (UPLC) on a Waters HPLC system using ACQUITY UPLC®BEH C18 1.7 μm (2.1*50mm column).

2. Color coordinates (L*, a*, and b*)

[0142] The color coordinates of the recycled bisphenol A monomer composition were analyzed in reflection mode using HunterLab UltraScan PRO Spectrophotometer.

3. Recovery rate of by-product (DEC)

[0143] The percentage ratio of the number of moles of recovered DEC to the number of moles of DEC in the diethyl carbonate (DEC) by-product mixture containing MC, EtOH, and water separated in the distillation step was determined according to the following mathematical formula.

[Mathematical Formula]

$$\text{Recovery rate of diethyl carbonate} = \{(\text{Number of moles of diethyl carbonate recovered}) / (\text{Number of moles of diethyl carbonate in entire by-product mixture})\} \times 100.$$

[0144] Specifically, the number of moles of DEC in the by-product mixture can be measured by measuring the number of moles of DEC produced through a gas chromatography-flame ionization detector (GC-FID) device for the mixed solution immediately after depolymerization. The number of moles of recovered DEC can be that in which the number of moles of a by-product solution was measured for DEC by the same method through gas chromatography-flame

ionization detector (GC-FID) device.

[Table 2]

Measurement result of Experimental Example 1					
Category	Purity (%)	L*	a*	b*	DEC Recovery rate (%)
Example 1	99.72	98.91	-0.06	1.49	89.2
Example 2	99.13	96.91	-0.01	1.53	88.1
Example 3	99.14	97.12	0.06	1.56	87.9
Example 4	97.29	96.19	0.33	2.21	85.1
Example 5	96.82	97.82	0.31	1.82	87.1
Example 6	99.85	98.82	-0.05	1.48	88.8
Example 7	96.13	97.11	0.02	1.55	88.1
Example 8	97.11	96.92	0.26	2.10	86.2
Example 9	97.85	96.98	0.21	1.62	88.5
Example 10	99.12	97.11	0.11	1.51	87.6
Comparative Example 1	96.12	96.01	0.11	2.62	81.2
Comparative Example 2	96.87	94.12	0.55	3.11	77.1
Comparative Example 3	96.97	95.11	0.45	2.79	79.2
Comparative Example 4	95.81	94.19	0.41	3.02	76.6

[0145] As shown in Table 1, the recycled bisphenol A monomer compositions obtained in Examples 1 to 10 exhibited high purities of 96.13% to 99.85%. Also, the recycled bisphenol A monomer compositions obtained in Examples 1 to 10 exhibited color coordinates L* of 96.19 to 98.91, a* of -0.06 to 0.33, and b* of 1.48-2.21, showing excellent optical properties. In addition, the recycled bisphenol A monomer compositions obtained in Examples 1 to 10 were measured to have high DEC recovery rates of 85.1% to 89.2%. On the other hand, the recycled bisphenol A monomer compositions obtained in Comparative Examples 1 to 4 exhibited purities of 95.81% to 96.97%, which was decreased as compared to Examples. Further, the recycled bisphenol A monomer compositions obtained in Comparative Examples 1 to 4 exhibited color coordinates L* of 94.12 to 96.01, a* of 0.11 to 0.55, and b* of 2.62 to 3.11, showing poor optical properties as compared to Examples. Furthermore, the recycled bisphenol A monomer compositions obtained in Comparative Examples 1 to 4 had DEC recovery rates of 76.6% to 81.2%, which were lower than those of Examples.

Claims

1. A monomer composition for synthesizing recycled plastic,

which comprises an aromatic diol compound,
wherein diethyl carbonate is obtained as a by-product,
wherein a recovery rate of diethyl carbonate, which is a by-product, according to the following mathematical formula is 82% or more, and
wherein the monomer composition for synthesizing recycled plastic is recovered from a polycarbonate-based resin,

[Mathematical Formula]

$$\text{Recovery rate of diethyl carbonate} = \{(\text{Number of moles of diethyl carbonate recovered}) / (\text{Number of moles of diethyl carbonate in entire by-product mixture})\} \times 100.$$

2. The monomer composition for synthesizing recycled plastic according to claim 1 wherein:
the monomer composition for synthesizing recycled plastic has a color coordinate L* of 96.1 or more.

3. The monomer composition for synthesizing recycled plastic according to claim 1 wherein:
the monomer composition for synthesizing recycled plastic has a color coordinate a* of 0.4 or less.

4. The monomer composition for synthesizing recycled plastic according to claim 1 wherein:
the monomer composition for synthesizing recycled plastic has a color coordinate b* of 2.5 or less.

5. The monomer composition for synthesizing recycled plastic according to claim 1 wherein:
the monomer composition for synthesizing recycled plastic has an aromatic diol compound purity of 96% or more.

6. The monomer composition for synthesizing recycled plastic according to claim 1 wherein:
the aromatic diol compound is recovered from the polycarbonate-based resin.

7. A method for preparing a monomer composition for synthesizing recycled plastic, the method comprising the steps of:

depolymerizing a polycarbonate-based resin;
adding a first adsorbent to a depolymerization reaction product to perform an adsorption purification and then removing the first adsorbent;
separating a carbonate precursor from the depolymerization reaction product; and
purifying the depolymerization reaction product from which the carbonate precursor has been separated,
wherein the step of purifying the depolymerization reaction product from which the carbonate precursor has been separated comprises: a step of washing the depolymerization reaction product from which the carbonate precursor has been separated; and a step of adding a second adsorbent to the depolymerization reaction product from which the carbonate precursor has been separated to perform an adsorption purification and then removing the second adsorbent.

8. The method for preparing a monomer composition for synthesizing recycled plastic according to claim 7 wherein:
the first adsorbent and the second adsorbent are the same or different from each other, and each independently include at least one activated carbon selected from the group consisting of a plant-based activated carbon, a coal-based activated carbon, a petroleum-based activated carbon, and a waste-based activated carbon.

9. The method for preparing a monomer composition for synthesizing recycled plastic according to claim 7 wherein:
an amount of the first adsorbent added is 1 part by weight to 1000 parts by weight based on 100 parts by weight of the second adsorbent added.

10. The method for preparing a monomer composition for synthesizing recycled plastic according to claim 7 wherein:
the washing step comprises a step of washing with a solvent at a temperature of 10°C or more and 30°C or less; and a step of washing with a solvent at a temperature of 40°C or more and 80°C or less.

11. The method for preparing a monomer composition for synthesizing recycled plastic according to claim 10 wherein:
the solvent of the step of washing with a solvent at a temperature of 10°C or more and 30°C or less is an organic solvent.

12. The method for preparing a monomer composition for synthesizing recycled plastic according to claim 10 wherein:
the solvent of the step of washing with a solvent at a temperature of 40°C or more and 80°C or less is water.

13. The method for preparing a monomer composition for synthesizing recycled plastic according to claim 10 wherein:
a difference value between the temperature of the step of washing with a solvent at a temperature of 40°C or more and 80°C or less, and the temperature of the step of washing with a solvent at a temperature of 10°C or more and 30°C or less is 20°C or more and 50°C or less.

14. The method for preparing a monomer composition for synthesizing recycled plastic according to claim 7 wherein:
the depolymerization reaction of the polycarbonate-based resin is carried out in the presence of a solvent containing ethanol.

15. The method for preparing a monomer composition for synthesizing recycled plastic according to claim 7 wherein:

the depolymerization reaction of the polycarbonate-based resin is carried out by reacting a base in an amount of 0.5 moles or less relative to 1 mole of the polycarbonate-based resin.

16. The method for preparing a monomer composition for synthesizing recycled plastic according to claim 7 wherein:

the step of separating a carbonate precursor from the depolymerization reaction product comprises, a reduced pressure distillation step of the depolymerization reaction product.

17. The method for preparing a monomer composition for synthesizing recycled plastic according to claim 7 wherein:

after the step of adding a second adsorbent to the depolymerization reaction product from which the carbonate precursor has been separated to perform an adsorption purification and then removing the second adsorbent, the purifying step further comprises a recrystallization step of the depolymerization reaction product from which the carbonate precursor has been separated.

18. The method for preparing a monomer composition for synthesizing recycled plastic according to claim 7, which further comprises a neutralization reaction step of the depolymerization reaction product with an acid, before the step of adding a first adsorbent to the depolymerization reaction product to perform an adsorption purification and then removing the first adsorbent.

19. A recycled plastic comprising a reaction product of the monomer composition for synthesizing recycled plastic of claim 1 and a comonomer.

20. A molded product comprising the recycled plastic of claim 19.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/KR2022/010309

A. CLASSIFICATION OF SUBJECT MATTER

C08G 64/30(2006.01)i; **C08J 11/24**(2006.01)i; **C07C 68/06**(2006.01)i; **C07C 7/10**(2006.01)i; **C07C 37/86**(2006.01)i; **C07C 7/12**(2006.01)i; **C07C 7/04**(2006.01)i; **C07C 67/54**(2006.01)i

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

C08G 64/30(2006.01); C07C 27/02(2006.01); C07C 37/00(2006.01); C07C 37/01(2006.01); C07C 37/055(2006.01); C07C 37/52(2006.01); C07C 68/00(2006.01); C08G 64/20(2006.01); C08G 64/38(2006.01); C08G 64/42(2006.01); C08J 11/24(2006.01)

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Korean utility models and applications for utility models: IPC as above
Japanese utility models and applications for utility models: IPC as above

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

eKOMPASS (KIPO internal) & keywords: 해중합(depolymerization), 폴리카보네이트(polycarbonate), 디올(diol), 디에틸 카보네이트(diethyl carbonate), 흡착제(absorbent), 정제(refining), 세척(wash), 재활용(recycling)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	KR 10-2015-0028359 A (SAUDI BASIC INDUSTRIES CORPORATION) 13 March 2015 (2015-03-13) See abstract; paragraphs [0002], [0010], [0036]-[0039], [0042], [0045]-[0047], [0074], [008 6] and [0094]; claims 1, 9, 15, 20 and 22; and example 4.	1-6,19,20
Y		7-9,14-18
A		10-13
Y	US 2015-0105531 A1 (SAUDI BASIC INDUSTRIES CORPORATION) 16 April 2015 (2015-04-16) See abstract; and paragraphs [0021], [0033] and [0090].	7-9,14-18
Y	JP 2004-277396 A (TEIJIN CHEM. LTD.) 07 October 2004 (2004-10-07) See abstract; and paragraph [0026].	18

☑ Further documents are listed in the continuation of Box C. ☑ See patent family annex.

* Special categories of cited documents:
"A" document defining the general state of the art which is not considered to be of particular relevance
"D" document cited by the applicant in the international application
"E" earlier application or patent but published on or after the international filing date
"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
"O" document referring to an oral disclosure, use, exhibition or other means
"P" document published prior to the international filing date but later than the priority date claimed
"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
31 October 2022	**01 November 2022**
Name and mailing address of the ISA/KR **Korean Intellectual Property Office** **Government Complex-Daejeon Building 4, 189 Cheongsa-ro, Seo-gu, Daejeon 35208** Facsimile No. **+82-42-481-8578**	Authorized officer Telephone No.

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INTERNATIONAL SEARCH REPORT

International application No.
PCT/KR2022/010309

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	JP 2006-022183 A (TEIJIN CHEM. LTD.) 26 January 2006 (2006-01-26) See entire document.	1-20
A	KR 10-2020-0014004 A (INDUSTRIAL COOPERATION FOUNDATION CHONBUK NATIONAL UNIVERSITY) 10 February 2020 (2020-02-10) See entire document.	1-20
A	JP 3422647 B2 (PROD. DEV. RES. INST.) 30 June 2003 (2003-06-30) See entire document.	1-20

Form PCT/ISA/210 (second sheet) (July 2022)

INTERNATIONAL SEARCH REPORT
Information on patent family members

International application No.
PCT/KR2022/010309

Patent document cited in search report	Publication date (day/month/year)	Patent family member(s)	Publication date (day/month/year)
KR 10-2015-0028359 A	13 March 2015	CN 104718238 A	17 June 2015
		CN 104718238 B	24 August 2016
		DE 112013006107 T5	17 September 2015
		JP 2015-533816 A	26 November 2015
		JP 5881909 B2	09 March 2016
		US 8680227 B1	25 March 2014
		WO 2014-099596 A1	26 June 2014
US 2015-0105531 A1	16 April 2015	CA 2927187 A1	23 April 2015
		CN 105658712 A	08 June 2016
		EP 3058019 A1	24 August 2016
		JP 2016-533416 A	27 October 2016
		US 2016-0168063 A1	16 June 2016
		US 9328046 B2	03 May 2016
		WO 2015-057682 A1	23 April 2015
JP 2004-277396 A	07 October 2004	None	
JP 2006-022183 A	26 January 2006	JP 4272123 B2	03 June 2009
KR 10-2020-0014004 A	10 February 2020	KR 10-2090680 B1	18 March 2020
JP 3422647 B2	30 June 2003	JP 10-259151 A	29 September 1998

Form PCT/ISA/210 (patent family annex) (July 2022)

REFERENCES CITED IN THE DESCRIPTION

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Patent documents cited in the description

- KR 1020210122001 **[0001]**
- KR 1020210122002 **[0001]**
- KR 1020210122003 **[0001]**
- KR 1020210122004 **[0001]**
- KR 1020210128892 **[0001]**
- KR 1020210136153 **[0001]**